Population inversion with respect to the ground state generated in recombination

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Introduction

The introduction of CPA high power short pulse lasers has opened a new area of development of recombination pumping for soft X-ray lasers. In particular it becomes feasible to envisage that the pump pulse may be made sufficiently short to allow laser action from the excited to the ground state despite the self-terminating nature of these transitions. The hydrogen-like sequence of ions are particularly attractive for this application, having a large energy gap between the ground and resonance (n=2) states. Using linearly polarized pump light ensures a population of cold electrons following ATI ionization, which can guarantee a completely fully stripped set of ions^[1]. Adding hydrogen buffer gas to increase the cold electron population gives a good measure of control over the effective electron density and temperature independent of lasant ion^[2].

Initial studies considered cases of lasing between excited states, but more recently attention has turned to transitions to the ground state^[3]. In a detailed study involving the temporal development of the electron distribution function resulting from inverse bremsstrahlung, electron equilibration and three-body recombination heating, the author found that the initial non-Maxwellian electron distribution assumed a quasi-Maxwellian form with a high energy tail, but a cold mode, which governed the recombination^[4]. Population inversions to the ground state were generated in the subsequent recombination cascade, but were relatively short lived.

In view of this encouraging result, it became worthwhile to re-examine the general theory of cascade recombination to see the conditions under which ground state inversions could be generated. This theory was developed during the 1960's using three different approaches. The most general model is the collsional-radiative approach of Bates *et al.* ^[5] and McWhirter and Hearn^[6], which has been used in most subsequent work; the Fokker-Planck approach^[7] which is valid in the limit of low temperature; and the heuristic 'bottleneck' picture^[8], which provides a useful visualization of the problem.

In this work we have used the collisional-radiative model with 200 excited levels specified by their principal quantum number, n, to represent a hydrogenic-ion of charge Z. The collisional excitation and ionization rates are calculated using the Bethe approximation (the latter by analytic continuation into the continuum from the former), and the empirical Gaunt factor^[9], which is adequate for this purpose. De-excitation and direct three-body recombination rates are given by detailed balance. Radiative rates are generated by the standard dipole transition rate with recombination again by extension into the continuum.

Depression of ionization is not included. The higher states provide the correct density of states and excitation/deexcitation rates as in the depressed continuum provided they are continuous across the ionization boundary as is the case here. Problems will arise if we self-consistently calculate the loss of electrons from the background when it will be important to carefully estimate the number in bound states. However as we are envisaging a large background electron density from the buffer gas, we may treat the electron density as constant. We will also neglect electron heating during three body recombination for the same reason.

Scaling relations

We cast our primary variables into dimensionless forms.

$$Y = \frac{I_{1}}{kT} = \frac{Z^{2}I_{H}}{kT}$$

$$N_{e} = \frac{h^{3}}{2(2\pi m_{e}kT)^{3/2}}n_{e}$$
(1)

and similar forms for the ion and excited state densities N_{∞} and N_n . Since the Fermi energy $E_F = (h^2/2m_e)(3n_e/\pi)^{2/3}$, the term $N_e = (1/6\sqrt{\pi})(E_F/kT)^{3/2}$ represents the condition for the avoidance of the onset of degeneracy, namely $N_e << 0.1$. I_n is the ionization energy of state n.

In terms of these variables we may write the basic equations to be solved for the time development of the state populations as

$$\frac{dq_n}{dt} = \sum_m C_{nm} q_m - \sum_m C_{mn} q_{mn} + \sum_{m>n} A_{nm} q_m - \sum_{m
(2)$$

where q_n is the population of the state n, q_{∞} the continuum population, and C_{nm} and A_{nm} the collisional and radiative rates from the state m to n. The continuum population is given by the equation

$$\frac{dq_{\infty}}{dt} = \sum_{n} \left[\mathcal{C}_{\infty n} q_{n} - \mathcal{C}_{n\infty} + A_{n\infty} \right] q_{\infty} \left[\right]$$
(3)

Summing we obtain the result that

$$\sum_{n} q_n + q_\infty = l$$

i.e. the total population is constant. The collisional rate coefficients are conveniently written in terms of a rate $A = 2I_H/h = 6.575 \times 10^{15} \text{ sec}^{-1}$, and the radiative constant $A \times B$ where $B = \pi \alpha^3 Z^4 = 1.221 \times 10^{-6} Z^4$ and α is the fine structure coefficient. In terms of these variables the time used in equations (2) and (3) is written in units of $(A N_e)^{-1}$.

In the steady state the analysis can be further reduced by a simple scaling with Z. From Saha's equation which in terms of (1) takes the form

$$\frac{N_e N_{\infty}}{N_n} = \frac{g_{\infty}}{g_n} exp\left(-\frac{I_n}{kT}\right)$$
(4)

it is clear that N_n and N_∞ must scale differently. Thus eqn.(2) cannot be satisfied. However in the steady state, we can neglect eqn.(2), set N_∞ to an appropriate value (unity) and calculate relative populations and rates accordingly. Thus for this situation we may introduce the scaling $N_e \sim Z^4$, and appropriate scalings for N_n and N_∞ ^[10], which satisfy eqn.(4).

Development of the excited state populations

We consider the case where the ions are initially fully stripped into bare nuclei and follow the development of the population in the excited states. Fig.1 shows a typical case.



Figure 1. Plots of the populations of the excited states scaled against the Saha population plotted for principal quantum numbers 1-20 for different times for the case Z=3, $N_e=3\times10^{-4}$ and Y=40.

The high lying states equilibrate with the continuum extremely rapidly, in a time $\sim Y/n^2$. As can be seen the populations progressively diffuse to their equilibrium value. By time 10 all states except n=2 have practically reached their steady state. There is correspondingly a significant reduction in the continuum population with most electrons in the high lying states – the normal thermal equilibration process continued into the initial void below the continuum.

The consequence of progressive filling of the excited states is the establishment of population inversions.

Fig.2 shows the growth of the population, plotted as q_n/g_n , in a lithium system following ionization at a temperature Y=40 (3eV.). It can be seen that at the higher density 3×10^{-4} (10^{19} cm⁻³) an inversion is formed between the ground state and the resonance level lasting for about 16 units (8ps.) However at the lower density 6×10^{-5} (2×10^{18} cm⁻³) no inversion is formed. Indeed it can be seen that this case is critical in that the n=1 and n=2 profiles touch.



Figure 2. Time development of the reduced populations of the ground, and first and second excited states in Li^{2+} at temperature Y=40 and two different densities $N_e=6\times10^{-5}$ and $N_e=3\times10^{-4}$.

Examining fig.2 more closely we can see that the history of the n=3 is the same at both densities, and the n=2 nearly so. Clearly this reflects a scaling of both the temporal development and the final steady state values of the population, which are independent of the density but vary with the temperature.



Figure 3. Time development of the reduced populations of the ground, and first and second excited states in C^{5+} at temperature Y=40 and densities $N_c=3.4\times10^{-3}$.

Fig.3 shows a similar threshold case in carbon at the same temperature Y=40 (12eV.) and density $N_e=3.4\times10^{-3}$ (9×10²⁰cm⁻³). It can be seen that in this case also the population graphs of n=1 and n=2 touch.

If we contrast the two limiting cases in figs 2 and 3 we observe that the time history in our reduced units is identical. This in fact is an example of a general rule, namely that the population development and values of the populations scale with the density parameter $Z^{-4} N_e$. This scaling is associated with the steady state picture and appears in violation of the conservation equation (3). It arises from a combination of factors

- 1. Most of the population lies in the high lying excited states and the continuum population is small, generating an approximately steady-state distribution amongst the excited states.
- 2. Introducing the Z⁻⁴ scaling to N_e and hence to the time removes the Z scaling of the radiation constant B.
- 3. The ground state population is small.

The key to the generation of the inversion can now be seen in that the 'bottleneck' at $n\approx\sqrt{Y}$ lies near to but above n=2so that the resonance level is strongly pumped by the states immediately above, predominantly by collision during the cascade. The ground state on the other hand is dominantly pumped by radiative transitions from the states immediately above it, n=2, 3 and a smaller contribution from the high lying states.

We can identify the condition for the onset of inversion in terms of a relationship between the density $Z^{\text{-}4}\,N_e$ and the temperature Y

| Y | 20 | 40 | 100 | 150 | |
|--------------------------------|----------|----------------------|----------------------|----------|--|
| Z ⁻⁴ N _e | 3.7×10-7 | 7.4×10 ⁻⁷ | 1.2×10 ⁻⁶ | 1.5×10-6 | |

Table 1. Limiting density parameter N_e in dimensionless units.

As the temperature of the plasma decreases (Y increasing) the value of N_e required to generate the inversion must increase. However the definition of N_e is itself quite strongly temperature dependent, eqn.(1), and the necessary electron density in fact decreases as can be seen from the data in Table 2. This reflects the strong increase in the collisional cascade recombination rate with decreasing temperature, whereas the radiative recombination rate to the ground state increases more slowly.

| Z ⁻² kT (eV). | 0.68 | 0.34 | 0.136 | 0.091 |
|---|-----------------------|-----------------------|-----------------------|-----------------------|
| Z ⁻⁷ ne (cm ⁻³) | 1.25×10 ¹⁵ | 8.86×10 ¹⁴ | 3.64×10 ¹⁴ | 2.47×10 ¹⁴ |

Table 2. Limiting density ne in practical units.

We note however the importance of the limit imposed by degeneracy at high Z and low temperature on the data in Table 1.

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